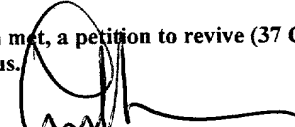


JCO7 Rec'd PCT/10 29 MAR 2001

FORM PTO-1390 (REV. 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER SPO-593	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (if known) CIP 1.5 09/806305	
INTERNATIONAL APPLICATION NO. PCT/JP00/03904		INTERNATIONAL FILING DATE 15 June 2000		PRIORITY DATE CLAIMED 18 August 1999	
TITLE OF INVENTION LAMINATED FILM AND METHOD OF PRODUCING THE SAME					
APPLICANT(S) FOR DO/EO/US Kenji MIHARU, et al.					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
<p>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.</p> <p>4. <input type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p style="margin-left: 20px;">a. <input type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau).</p> <p style="margin-left: 20px;">b. <input checked="" type="checkbox"/> has been communicated by the International Bureau.</p> <p style="margin-left: 20px;">c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</p> <p>6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).</p> <p style="margin-left: 20px;">a. <input checked="" type="checkbox"/> is attached hereto.</p> <p style="margin-left: 20px;">b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).</p> <p>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p style="margin-left: 20px;">a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau).</p> <p style="margin-left: 20px;">b. <input type="checkbox"/> have been communicated by the International Bureau.</p> <p style="margin-left: 20px;">c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p style="margin-left: 20px;">d. <input checked="" type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).</p> <p>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p> <p>Items 11 to 20 below concern document(s) or information included:</p> <p>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment.</p> <p>14. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>15. <input type="checkbox"/> A substitute specification.</p> <p>16. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.</p> <p>18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</p> <p>19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).</p> <p>20. <input checked="" type="checkbox"/> Other items or information: Copy of PCT/IB/308</p>					

U.S. APPLICATION NO. (known, see 37 CFR 1.53) 09/806305		INTERNATIONAL APPLICATION NO. PCT/JP00/03904		ATTORNEY'S DOCKET NUMBER SPO-593	
21. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO. \$1000.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$860.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00 ENTER APPROPRIATE BASIC FEE AMOUNT =				CALCULATIONS PTO USE ONLY <div style="border: 1px solid black; padding: 2px; display: inline-block;">\$ 860.00</div>	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				<div style="border: 1px solid black; padding: 2px; display: inline-block;">\$ 0</div>	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	
Total claims	8 - 20 =	0	x \$18.00	\$ 0	
Independent claims	3 - 3 =	0	x \$80.00	\$ 0	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$270.00	\$ 0	
TOTAL OF ABOVE CALCULATIONS =				\$ 860.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				+ \$ 0	
SUBTOTAL =				\$ 860.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$ 0	
TOTAL NATIONAL FEE =				\$ 860.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				\$ 40.00	
TOTAL FEES ENCLOSED =				\$ 900.00	
				Amount to be refunded: \$	
				charged: \$	
a. <input checked="" type="checkbox"/> A check in the amount of \$ <u>900.00</u> to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>19-1980</u> . A duplicate copy of this sheet is enclosed. d. <input type="checkbox"/> Fees are to be charged to a credit card. WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO <div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> SHERMAN & SHALLOWAY 413 N. Washington Street Alexandria, VA 22314 703-549-2282 </div> <div style="width: 45%; text-align: right;"> <div style="text-align: center; margin-bottom: 10px;">  SIGNATURE </div> <div style="text-align: center; margin-bottom: 10px;"> <u>Leonard W. Sherman</u> NAME </div> <div style="text-align: center;"> <u>19,636</u> REGISTRATION NUMBER </div> </div> </div>					

09/806305

SPO-593

JC08 Rec'd PCT/PTO PATENT
29 MAR 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of:

Kenji MIHARU, et al.

Serial No.: Unassigned

Group:

Filed: Concurrently

Examiner:

FOR: LAMINATED FILM AND METHOD OF PRODUCING THE SAME

Date: March 29, 2001

The Hon. Commissioner of
Patents and Trademarks
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Sir:

Preliminary to examination, please amend the herewith filed
application as follows:

IN THE CLAIMS

Please cancel Claim 5 in its entirety and without
prejudice.

Please add the following new claims:

--8. (New) A laminated film according to claim 1,
wherein another polar base member is laminated on the polyester
film via the extrusion-laminated resin.

9. (New) A laminated film according to claim 2, wherein

09806305-03901

another polar base member is laminated on the polyester film via the extrusion-laminated resin.--

REMARKS

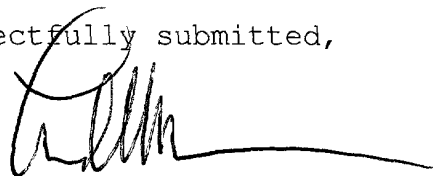
Entry of the foregoing amendment prior to examination of this application is respectfully requested in view of the following comments.

Claim 5 has been cancelled and new claims 8 and 9 have been added. Accordingly, claims 1-4 and 6-9 are pending in this application.

Claim 5 has been cancelled to delete the multiple dependency therein. New claims 8 and 9 correspond to the multiple dependency rewritten as single dependent claims.

No new matter has been added and applicant respectfully submits that this application is in condition for allowance and an early notice to that effect is earnestly solicited.

Respectfully submitted,



Attorney for Applicants
Leonard W. Sherman
Reg. No. 19,636

SHERMAN & SHALLOWAY
P.O. BOX 788
Alexandria, Virginia 22313
(703) 549-2282

09306305-032901

SPECIFICATION

LAMINATED FILM AND METHOD OF PRODUCING THE SAME

(Technical Field)

- 5 The present invention relates to a polyester
laminated film obtained by extrusion-laminating an
ethylene copolymer resin onto a polyester film, exhibiting
excellent interlayer adhesiveness to the polyester
surface, and to a method of producing the same. More
10 specifically, the invention relates to a method of
producing a polyester laminated film having excellent
interlayer adhesiveness by the extrusion-lamination
without effecting the anchor-coat treatment on the
polyester film or without effecting the ozone treatment on
15 the laminated resin film, and to a laminated film obtained
by the above method.

(Background Art)

- A polyester film and, particularly, a biaxially
stretched polyester film exhibits excellent transparency,
20 toughness and oxygen barrier property, and has been widely
used as a base member of packaging materials for packaging
foods and industrial materials. In practice, other base
member that plays the roll of a reinforcing member, such
as polyamide film, ethylene/vinyl alcohol copolymer,
25 aluminum foil, aluminum-deposited polyester film,
inorganic material-deposited polyester film or paper, is
further laminated on the polyester film and, besides, a
heat-sealing resin layer is laminated thereon to impart
heat-sealing property.

- 30 The films can be laminated by such a laminating
method as dry-laminating method, wet-laminating method or
extrusion-laminating method. Among them, the extrusion-
laminating method is widely used which is most
advantageous from the standpoint of cost of production and
35 efficiency. Here, the lamination of the polyester film on

It has therefore been strongly urged in the field of packaging industries to provide a method of producing a polyester laminated film having favorable interlayer
35 adhesiveness by the extrusion-laminating method without

5 discovered the fact that use of the anchor-coating agent and the ozone-treating apparatus can be omitted when a resin having a particular composition is extrusion-laminated on the polyester film, and have arrived at the present invention.

It is therefore an object of the present invention to provide a method of producing a laminated film having excellent interlayer adhesiveness based on the extrusion-lamination method by laminating the polyester films or by
15 laminating the polyester film on another base member film, and a laminated film obtained by this method.

The invention is further concerned with a laminated
30 film obtained by extrusion-laminating, onto at least one
surface of a polyester film, a mixture resin comprising:

(a) 100 parts by weight of the mixture resin component of an ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer or the mixture
35 resin composition of said ethylene/unsaturated carboxylic

acid/(meth)acrylic acid ester copolymer with an ethylene/unsaturated carboxylic acid copolymer and/or an ethylene/(meth)acrylic acid ester copolymer, the amount of the unsaturated carboxylic acid component being from 1 to 12% by weight and the amount of the (meth)acrylic acid ester component being from 2 to 25% by weight with respect to the total amount of the extrusion-laminated resin components, and

(b) not more than 30 parts by weight of an ethylene/ α -olefin copolymer resin having a density of 840 to 900 kg/cm³.

The invention further relates to a method of producing the laminated film by extrusion-laminating, at a resin temperature of 280 to 340 °C, the above extrusion-laminate resin onto a polyester of which the surface has been oxidized and having a surface wet tension of not smaller than 45 dyns/cm.

(Detailed Description of the Invention)

It is desired that a polyester constituting the polyester film used in the present invention chiefly contains, as an acid component, an aromatic dicarboxylic acid and, particularly, a terephthalic acid or a 2,6-naphthalenedicarboxylic acid in an amount of not smaller than 80 mol% and, preferably, not smaller than 90 mol% from the standpoint of transparency, heat resistance, toughness and gas barrier property. As the acid component, there may be further contained other aromatic dicarboxylic acid, such as isophthalic acid, phthalic acid, as well as an alicyclic dicarboxylic acid such as 1,4-cyclohexanedicarboxylic acid or an aliphatic dicarboxylic acid such as adipic acid or sebacic acid as a copolymerizable component. There may be further added trifunctional or more functional carboxylic acids such as trimellitic acid, hemimellitic acid and pyromellitic acid as copolymerizable compounds provided their amounts are

small.

As the dihydroxyl compound component constituting the polyester, there can be exemplified an aliphatic glycol such as an ethylene glycol, a propylene glycol, a 1,4-
5 butanediol, a 1,6-hexanediol, a neopentyl glycol, a diethylene glycol and a triethylene glycol, an alicyclic diol such as 1,4-cyclohexanediol, and an aromatic dihydroxyl compound such as bisphenol A ethylene oxide adduct. Among them, it is desired to use the one
10 containing ethylene glycol or 1,4-butanediol in an amount of not smaller than 80 mol% and, preferably, not smaller than 90 mol%. There may be further contained trifunctional or more functional hydroxyl compound such as glycerin, trimethylolpropane or the like provided their
15 amounts are small.

As representative polyesters, there can be exemplified polyethylene terephthalate, polyethylene-2,6-naphthalene dicarboxylate and polybutylene terephthalate.

It is further desired that the polyester has an
20 intrinsic viscosity of from 0.5 to 1.4 dl/g and, particularly, from 0.6 to 1.0 dl/g as measured at 20 °C and at a concentration of 0.4 g/100 ml in a 47/53 (volume ratio) mixture solvent of trifluoroacetic acid/methane dichloride. It is further desired that the polyester has
25 a melting point (temperature exhibiting a maximum endothermic peak as measured by using a differential scanning calorimeter) of not lower than 210 °C and, particularly, not lower than 220 °C.

The polyester film can be industrially produced as a
30 single-layer film of polyester by the known T-die cast film method or as a co-extruded film of two or more polyesters or by using other resin than the polyester by the T-die cast film method of the co-extrusion type. They may not be stretched, but are monoaxially stretched or
35 biaxially stretched to enhance the adaptability as the

09806305 03501
TOP SECRET 60E90860

base material film of the packaging material, and are available in the market. It is, however, desired to use the biaxially stretched films since they exhibit markedly improved toughness, transparency, heat resistance and gas barrier property.

In the present invention, the resin to be extrusion-laminated on the polyester film is an ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer or a mixture resin composition thereof with an ethylene/unsaturated carboxylic acid copolymer and/or an ethylene/(meth)acrylic acid ester copolymer, the amount of the unsaturated carboxylic acid component being from 1 to 12% by weight and the amount of the (meth)acrylic acid ester component being from 2 to 25% by weight with respect to the total amount of the resin components. That is, either the ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer is used alone or a mixture resin thereof with the ethylene/unsaturated carboxylic acid copolymer and/or the ethylene/(meth)acrylic acid ester copolymer is used. In either case, the unsaturated carboxylic acid component is contained in an amount of from 1 to 12% by weight and, preferably, from 2 to 6% by weight, and the (meth)acrylic acid ester component is contained in an amount of from 2 to 25% by weight and, preferably, from 6 to 20% by weight, as resins to be extrusion-laminated.

When the amount of the unsaturated carboxylic acid component is smaller than the above-mentioned range, the extrusion-laminated resin loses the polarity and poorly adheres to other polar base member such as polyester film or aluminum foil, and the utility of the laminated film is adversely affected. When the amount of the unsaturated carboxylic acid exceeds the above-mentioned range, on the other hand, the resin loses heat resistance, the unmolten products forms due to the dehydration crosslinking

reaction during the extrusion-laminate processing, and
foams develop conspicuously due to water. When the amount
of the (meth)acrylic acid ester component is smaller than
the above-mentioned range, affinity decreases between the
5 extrusion-laminated resin and the polyester film, and the
adhesiveness to the polyester decreases. When the amount
of the (meth)acrylic acid ester component exceeds the
above-mentioned range, on the other hand, the stickiness
increases during the extrusion-laminate processing,
10 deteriorating the adaptability for processing.

As the unsaturated carboxylic acid in the
ethylene/unsaturated carboxylic acid/(meth)acrylic acid
ester copolymer, there can be exemplified acrylic acid,
methacrylic acid, ethacrylic acid, fumaric acid, maleic
15 acid, monomethyl maleate and maleic anhydride. Among
them, it is particularly desired to use a copolymer of
acrylic acid or methacrylic acid. The (meth)acrylic acid
ester in the copolymer stands for an acrylic acid ester or
a methacrylic acid ester. Concrete examples include
20 esters of such as methyl, ethyl, n-butyl, isobutyl, 2-
ethylhexyl, and isooctyl. In the copolymer, the
unsaturated carboxylic acid is contained in an amount of,
desirably, from 1 to 15% by weight and, particularly, from
2 to 12% by weight, and the (meth)acrylic acid ester is
25 contained in an amount of from 4 to 25% by weight and,
particularly, from 6 to 20% by weight. It is further
desired that the copolymer has a melt flow rate (MFR) of
from 0.5 to 100 g/10 min. and, particularly, from 1 to 50
g/10 min. at 190 °C under a load of 2160 g.

30 As the ethylene/unsaturated carboxylic acid copolymer
that can be used together with the ethylene/unsaturated
carboxylic acid/(meth)acrylic acid ester copolymer, there
can be exemplified copolymers of ethylene with acrylic
acid, methacrylic acid, ethacrylic acid, fumaric acid,
35 maleic acid, monomethyl maleate or maleic anhydride.

Among them, it is particularly desired to use an ethylene/acrylic acid copolymer or an ethylene/methacrylic acid copolymer. It is desired that the content of the unsaturated carboxylic acid in the copolymer is from 1 to 5 15% by weight and, particularly, from 2 to 12% by weight. It is further desired that the copolymer has an MFR of from 0.5 to 100 g/10 min. and, particularly, from 1 to 50 g/10 min. at 190 °C under a load of 2160 g.

As the ethylene/(meth)acrylic acid ester copolymer 10 that can be used together with the ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer, there can be exemplified a copolymer of an ethylene and a (meth)acrylic acid ester. As the ester component, there can be exemplified esters of such as methyl, ethyl, n- 15 butyl, isobutyl, 2-ethylhexyl and isooctyl. It is further desired that the copolymer has an MFR of from 0.5 to 100 g/10 min. and, particularly, from 1 to 50 g/10 min. at 190 °C under a load of 2160 g.

These ethylene copolymers are obtained by the radical 20 copolymerization at a high temperature and under an elevated pressure.

In the second invention, the resin to be extrusion-laminated is a mixture resin comprising:

(a) 100 parts by weight of the mixture resin 25 component of an ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer or the mixture resin composition of said ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer with an ethylene/unsaturated carboxylic acid copolymer and/or an 30 ethylene/(meth)acrylic acid ester copolymer, the amount of the unsaturated carboxylic acid component being from 1 to 12 % by weight and the amount of the (meth)acrylic acid ester component being from 2 to 25 % by weight with respect to the total amount of the extrusion-laminated 35 resin components, and

03005305 032901
T0520 50590850

(b) not more than 30 parts by weight and preferably, from 5 to 20 parts by weight, and more preferably, from 10 to 20 parts by weight, of an ethylene/ α -olefin copolymer resin having a density of 840 to 900 kg/m³.

5 The ethylene/ α -olefin copolymer is a random copolymer of an ethylene and an α -olefin, such as propylene, 1-butene, 1-pentene, 1-hexene, 1-octene, 1-decene or 4-methyl-1-pentene. As such a copolymer, it is desired to use a copolymer of an ethylene and an α -olefin
10 having about 4 to about 8 carbon atoms. Further, the copolymer has a density of from 840 to 900 kg/m³ and, desirably, from 860 to 890 kg/m³.

 The ethylene/ α -olefin copolymer is used for enhancing the adhesive force to the polyester film. When
15 the density exceeds the above-mentioned range, however, the adhesion to the polyester film is not so much improved. When the density becomes smaller than the above range, on the other hand, the resin becomes so sticky that the handling is deteriorated during the extrusion-
20 lamination processing. When the blended amount exceeds the above-mentioned range, further, the extrusion-laminate processability greatly decreases.

 It is further desired that the copolymer has an MFR of from 0.5 to 200 g/10 min. and, particularly, from 1 to
25 50 g/10 min. at 190 °C under a load of 2160 g. Such a copolymer can be produced by using a combination catalyst of, for example, a transition metal catalyst component such as vanadium compound, zirconium compound or metallocene compound and an organic aluminum compound
30 catalyst component.

 The second invention is the same as the first invention except that the amount of the unsaturated carboxylic acid component is from 1 to 12% by weight and, preferably, from 2 to 6% by weight and the amount of the
35 (meth)acrylic acid ester component is from 2 to 25% by

weight and, preferably, from 6 to 20% by weight based on the ethylene/polar monomer copolymer in the extrusion-laminated resin.

In the first and second inventions, it is desired
5 that the extrusion-laminated resin has been so adjusted as to exhibit an MFR of from 0.5 to 100 g/10 min. and, particularly, from 1 to 50 g/10 min. at 190 °C under a load of 2160 g by taking the extrusion-laminate processability into consideration.

10 Further, the carboxylic acid group of the unsaturated carboxylic acid in the extrusion-laminated resin may be partly ionized with an alkali metal or an alkaline earth metal within a range in which the ionization degree is not larger than 20% and, particularly, not larger than 10%.
15 When the ionization degree exceeds 20%, the adhesive force to the polyester film decreases, which is not desirable.

If necessary, further, the resin to be extrusion-laminated may be blended with various additives, such as antioxidant, heat stabilizer, slipping agent and anti-
20 blocking agent. Concretely speaking, as the slipping agent or the anti-blocking agent, there can be used one or two or more of amides of saturated fatty acid or unsaturated fatty acid, such as stearic acid amide, oleic acid amide, erucic acid amide, palmitic acid amide, oleyl
25 palmitamide, stearyl erucamide, N,N'-ethylenebis erucamide, hydrogenated castor oil and silica. It is desired that they are used in an amount of from 0.1 to 5 parts by weight and, particularly, from 0.3 to 3 parts by weight per 100 parts by weight of the extrusion-lamination
30 resin.

The laminated film according to the first invention and the second invention can be produced by using a known extrusion laminator. Described below in detail is a method of producing a laminated film of a two-layer
35 structure comprising a biaxially stretched polyester

film/extrusion-laminated resin, which is the simplest constitution of the invention and is a representative embodiment.

The biaxially stretched polyester film is delivered
5 at a constant speed from the base member film delivery
portion and is guided to a laminating portion. In the
laminating portion, a thin molten film of the to-be-
laminated resin heated and melted at a predetermined
temperature in the cylinder and is continuously extruded
10 in the form of a thin film from the T-die, is permitted to
be hanged directly on the surface of the biaxially
stretched polyester film, and the biaxially stretched
polyester film and the laminated resin are press-adhered
together and cooled simultaneously between the cooling
15 roll and the press-adhering roll, and are then taken up as
a product in the take-up portion.

It is desired that the to-be-laminated surface of the
biaxially stretched polyester film has a surface wet
tension of not smaller than 45 dyns/cm and, more
20 preferably, not smaller than 50 dyns/cm for producing a
sufficiently large adhesive force to the extrusion-
laminated resin. When the surface wet tension is smaller
than 45 dyns/cm, the adhesive force is not sufficient. In
order for the surface wet tension of the biaxially
25 stretched polyester film to assume the above-mentioned
level, the surface of the film may be oxidized relying
upon a known industrial method such as corona treatment or
flame treatment. The surface oxidation treatment may be
executed in a step of producing the biaxially stretched
30 polyester film or just before the lamination in the step
of extrusion-lamination.

It is desired that the extrusion temperature during
the extrusion-lamination processing is from 280 to 340 °C
and, particularly, from 300 to 330 °C as the resin
35 temperature measured just under the T-die. When the resin

temperature is lower than 280 °C, the adhesive force is not sufficient between the laminated resin and the polyester film. When the resin temperature is higher than 340 °C, on the other hand, the laminated resin is

5 thermally deteriorated to a conspicuous degree.

The two layer-laminated film comprising the polyester film and the extrusion-laminated resin exhibits excellent interlayer adhesiveness to the polyester film owing to the laminated resin, as well as excellent heat-sealing

10 property and hot-tacking property, and can be extensively used as a film for pillow packaging for packaging foods and industrial materials, and as plastic films, sheets, and as a film for thermal lamination on the sheets, papers, metals, woods, etc.

15 In the laminated film of the invention, other polar member that plays the role of reinforcement can be further laminated on the polyester film via the laminated resin. As the polar base member, there can be exemplified those members having metallic or ceramic surfaces, such as

20 aluminum foil, aluminum-deposited polyester film, aluminum-deposited polypropylene film, silica-deposited polyester film, and alumina-deposited polyester film, as well as plastic films and papers, such as polyamide, ethylene/vinyl alcohol copolymer, etc.

25 When the polar base member is to be further laminated, there can be employed the so-called extrusion sandwich-lamination method according to which the polyester film is fed from the delivering portion to the laminating portion like in the above-mentioned extrusion-

30 lamination method, the laminated resin is stacked on the polyester film and, at the same time, the polar base member film is fed even from the side opposite to the surface where the laminated resin and the polyester film are laminated one upon the other, so as to be laminated on

35 the laminated resin. According to the sandwich-lamination

method, contrary to the above-mentioned procedure, the polar base member is fed from the delivering portion to the laminating portion, the extrusion-laminated resin is stacked on the polar base member and, at the same time, the polyester film is fed from the side opposite to the surface where the laminated resin and the polar base member are laminated one upon the other, so as to be laminated on the laminated resin, which is the extrusion sandwich-lamination method.

Described below are representative examples of laminations of the polyester film and various polar base members via the extrusion-laminated resin. Here, the extrusion-laminated surface of the polar base member may, as required, be oxidized. Usually, however, the laminated resin in the present invention tends to exhibit favorable extrusion-lamination strength for the polar base member without treatment. It is therefore sufficient to install the surface oxidation treatment facility only on the side of the extrusion-laminated surface of the biaxially stretched polyester film.

- (a) O-PET/EX/aluminum foil
- (b) O-PET/EX/aluminum-deposited polyester
- (c) O-PET/EX/aluminum-deposited polypropylene
- (d) O-PET/EX/alumina-deposited polyester
- (e) O-PET/EX/ethylene-vinyl alcohol copolymer
- (f) O-PET/EX/paper

Here, O-PET stands for a biaxially stretched polyester film, and EX stands for an extrusion-laminated resin.

If necessary, further, there may be laminated any other base material film and, for this purpose, there may be employed any known method such as the extrusion laminator that is repetitively used, the tandem laminator or the dry laminator, to obtain a desired laminated film.

There is no particular limitation on the thickness of

the laminated film. Usually, however, the polyester film has a thickness of from 1 to 1000 μm and the extrusion-laminated resin layer has a thickness of from 3 to 300 μm . (EXAMPLES)

- 5 Described below are Examples and Comparative Examples for explaining the effect of the present invention. The starting laminated resin materials used in Examples and in Comparative Examples, method of producing laminated films, and method of evaluating the laminated films were as
10 described below.

1. Starting resin materials to be laminated.

(A-1) Ethylene/methacrylic acid/isobutyl acrylate copolymer ①.

Content of methacrylic acid: 4% by weight, content of
15 isobutyl acrylate: 15% by weight, MFR: 27 g/10 min.

(A-2) Ethylene/methacrylic acid/isobutyl acrylate copolymer ②.

Content of methacrylic acid: 11% by weight, content of isobutyl acrylate: 8% by weight, MFR: 10 g/10 min.

20 (B-1) Ethylene/methacrylic acid copolymer ①.

Content of methacrylic acid: 4% by weight, MFR: 7 g/10 min.

(B-2) Ethylene/acrylic acid copolymer ②.

Content of acrylic acid: 5% by weight, MFR: 8 g/10
25 min.

(C-1) Ethylene/ α -olefin copolymer ①.

α -olefin: 1-butene, density: 885 kg/m³, MFR: 3.6 g/10 min.

(C-2) Ethylene/ α -olefin copolymer ②.

α -olefin: 1-butene, density: 860 kg/m³, MFR: 35 g/10
30 min.

(C-3) Ethylene/ α -olefin copolymer ③.

α -olefin: 1-octene, density: 870 kg/m³, MFR: 35 g/10 min.

35 (D-1) Polyethylene ①

Density: 917 kg/m³, MFR 7.0 g/10 min.

(E-1) Ethylene/ethyl acrylate copolymer ①

Content of ethyl acrylate: 9% by weight, MFR: 5 g/10 min.

5 2. Method of producing laminated films.

2-1. Constitutions of laminated films.

Lamination constitution ①: O-PET/laminated resin (25 μ m)

Lamination constitution ②: O-PET/laminated resin (25 μ m)/base member film (a)

10 Lamination constitution ③: O-PET/laminated resin (25 μ m)/base member film (b)

O-PET: Biaxially stretched polyester film (12 μ m),
P60 (one surface is corona-treated) produced
by Toray Co.

15 Base member film (a): aluminum foil (7 μ m)/biaxially
stretched polyester film (12 μ m)

Base member film (b): polyethylene (20 μ m)/anchor-
coating agent/biaxially
20 stretched polyester film (12 μ m).

2-2. Method of extrusion-lamination.

Resin pellets prepared to have a predetermined
composition were melt-extruded onto the biaxially
25 stretched polyester film by using an extrusion laminator,
to prepare laminated films having the above-mentioned
lamination constitutions ①, ② and ③ under the following
production conditions.

Extrusion laminator: 65 mm-diameter extruder,
30 opening width of T-die, 500 mm.

Extrusion laminator processing rate: 80 m/min.

Extruded resin temperature: 270 to 350 °C (resin
temperature just under the T-die is
measured using a contact-type
35 thermometer).

0906305 02501
T062E0 50E90360

Air gap: 120 mm

In the case of the lamination constitution ①, the biaxially stretched polyester film was delivered, and the resin to be laminated was laminated by extrusion. In the

- 5 cases of the lamination constitutions ② and ③, the biaxially stretched polyester film was delivered, and the resin to be laminated was sandwich-laminated on the base member films (a) and (b), respectively.

3. Items to be evaluated and methods of evaluation.

- 10 3-1. Extrusion-lamination processability of the laminated resin.

The stability of the laminated film during the extrusion-laminate processing in 2-2 above was evaluated by naked eyes. In this observation by eyes, there were
15 maintained the sampling conditions of laminated films at a processing rate of 80 m/min. and a thickness of extruded laminate of 25 μ m, and the extrusion conditions of a processing rate of 80 m/min. and a thickness of the extruded laminate of 25 μ m. A maximum processing rate was
20 evaluated while gradually increasing the processing rate only. The target of extrusion-lamination processability was set to be the maximum processing rate of not smaller than 200 m/min, and the processing rates of lower than this value were judged to be of not practicable levels.

- 25 3-2. Adhesiveness between the polyester film and the laminated resin.

An extrusion-laminated sample (lamination constitution ③) two days after the production was measured under the conditions of a sample width of 15 mm,
30 a peeling angle of 90 degrees and a tension speed of 300 mm/min. The degree of oxidation on the laminated surface of the polyester film was judged relying upon a measured value of surface wet tension. The surface wet tension was measured by using a wet tension measuring solution
35 (produced by Wako Junyaku Co.). A target value of the

adhesion strength was set to be 3 N/15 mm, and the adhesion strengths smaller than this value were judged to be of not practicable levels.

- 3-3. Adhesiveness between the aluminum foil and the
5 laminated resin.

An extrusion-laminated sample (lamination constitution ②) one day after the production was measured under the conditions of a sample width of 15 mm, a peeling angle of 90 degrees and a tension speed of 300 mm/min. A
10 target value of the adhesion strength was set to be 2 N/15 mm, and the adhesion strengths smaller than this value were judged to be of not practicable levels.

- 3-4. Heat-sealing property of the laminated film.

The laminated resin surfaces of the samples
15 (lamination constitution ①) were heat-sealed by using a heat sealer to measure the heat-sealing strength. The heat sealer was a one surface-heated bar sealer (produced by Toyo Seiki co.), and the sealing pressure was 0.2 MPa (real pressure) and the sealing time was 0.5 seconds. A
20 maximum target strength of the heat sealing was set to be not smaller than 30 N/15 mm, and strengths smaller than this value were judged to be of not practicable levels. (Example 1)

By using a resin composition of (A-1)
25 ethylene/methacrylic acid/isobutyl acrylate copolymer ① and (B-1) ethylene/methacrylic acid copolymer ① blended at a ratio shown in Table 1 as a resin to be extrusion-laminated, there were prepared the lamination constitutions ①, ② and ③ by the method of producing the
30 laminated film described in 2. above, and were evaluated for their extrusion-lamination processability, adhesive force to the polyester film, adhesive force to the aluminum foil and heat-sealing strength according to the items to be evaluated and methods of evaluation described
35 in 3. above.

In conducting the extrusion-lamination processing, the resin temperature was set to 300 °C, and the above resin composition was extrusion-laminated on the surface of the polyester film of the side corona-treated. The results were as shown in Table 1.

(Example 2)

The laminated film was prepared according to the same method and under the same conditions as those of Example 1 but changing the resin temperature during the extrusion-lamination processing to 320 °C, and the extrusion-lamination processability and the physical properties were evaluated. The results were as shown in Table 1.

(Comparative Example 1)

The laminated film was prepared according to the same method and under the same conditions as those of Example 1 but changing the resin temperature during the extrusion-lamination processing to 270 °C, and was evaluated. The obtained laminated film exhibited an adhesive force to the polyester film of as very low as 0.1 N/15 mm. The heat-sealing strength did not reach the practicable level, and the laminated film was judged to be not suited as a packaging material. The results were as shown in Table 1.

(Comparative Example 2)

The extrusion-lamination processing was conducted according to the same method and under the same conditions as those of Example 1 but changing the resin temperature during the extrusion-lamination processing to 350 °C. In this case, however, the resin composition developed foaming during the extrusion-lamination processing, and the laminated film of the molten state was perforated. Therefore, the laminated film could not be sampled and could not be evaluated. The results were as shown in Table 1.

(Comparative Example 3)

The laminated film was prepared according to the same

method and under the same conditions as those of Example 1 but laminating the polyester film on the surface of the side not treated with corona (surface wet tension of 44 dyns/cm) and changing the resin temperature to 320 °C, and its physical properties were evaluated. The obtained laminated film exhibited an adhesive force to the polyester film of as very low as 0.8 N/15 mm. The heat-sealing strength did not reach the practicable level, and the laminated film was judged to be not suited as a packaging material. The results were as shown in Table 1. (Example 3)

A laminated film was prepared according to the same method and under the same conditions as those of Example 1 but using (A-2) ethylene/methacrylic acid, isobutyl acrylate copolymer ② alone as the resin to be extrusion-laminated, and its physical properties were evaluated. The results were as shown in Table 1. (Example 4)

By using a resin composition of (A-2) ethylene/methacrylic acid/isobutyl acrylate copolymer ② and (C-1) ethylene/ α -olefin copolymer ① blended at a ratio shown in Table 1 as a resin to be extrusion-laminated, there were prepared the lamination constitutions ①, ② and ③ by the method of producing the laminated film described in 2. above, and were evaluated for their extrusion-lamination processability, adhesive force to the polyester film, adhesive force to the aluminum foil and heat-sealing strength according to the items to be evaluated and methods of evaluation described in 3. above.

In conducting the extrusion-lamination processing, the resin temperature was set to 285 °C, and the above resin composition was extrusion-laminated on the surface of the polyester film of the side corona-treated. The results were as shown in Table 2.

(Example 5)

A laminated film was prepared according to the same method and under the same conditions as those of Example 4 but using (C-2) ethylene/ α -olefin copolymer ② instead of the component (C-1) as the resin to be extrusion-laminated and changing the resin temperature during the extrusion-lamination processing to 300 °C, and was evaluated. The results were as shown in Table 2.

(Example 6)

A laminated film was prepared according to the same method and under the same conditions as those of Example 4 but using (C-3) ethylene/ α -olefin copolymer ③ instead of the component (C-1) as the resin to be extrusion-laminated and changing the resin temperature during the extrusion-laminate processing to 300 °C, and was evaluated. The results were as shown in Table 2.

(Comparative Example 4)

The laminated film was prepared according to the same method and under the same conditions as those of Example 5 but laminating the polyester film on the surface of the side not treated with corona (surface wet tension of 44 dyns/cm), and was evaluated. The obtained laminated film exhibited an adhesive force to the polyester film of as very low as 0.7 N/15 mm. The heat-sealing strength did not reach the practicable level, and the laminated film was judged to be not suited as a packaging material. The results were as shown in Table 2.

(Example 7)

Properties were evaluated by according to the same method and under the same conditions as those of Example 1 but using a resin composition of (A-1) ethylene/methacrylic acid/isobutyl acrylate copolymer ①, (B-1) ethylene/methacrylic acid copolymer ① and (C-1) ethylene/ α -olefin copolymer ① blended at a ratio shown in Table 2 as a resin to be extrusion-laminated and

changing the resin temperature to 315 °C. The results were as shown in Table 2.

(Comparative Example 5)

The laminated film was prepared according to the same method and under the same conditions as those of Example 7 but laminating the polyester film on the surface of the side not treated with corona (surface wet tension of 44 dyns/cm), and was evaluated. The obtained laminated film exhibited an adhesive force to the polyester film of as very low as 0.6 N/15 mm. The heat-sealing strength did not reach the practicable level, and the laminated film was judged to be not suited as a packaging material. The results were as shown in Table 2.

(Example 8)

Properties were evaluated according to the same method and under the same conditions as those of Example 1 but using a resin composition of (A-2)

ethylene/methacrylic acid/isobutyl acrylate copolymer ② and (C-2) ethylene/ α -olefin copolymer ② blended at a ratio shown in Table 3 as a resin to be extrusion-laminated, delivering the surface of the polyester film which has not been treated with corona as the surface to be laminated, and executing the extrusion-lamination by effecting the corona-treatment using a corona-treating apparatus installed in the laminator just prior to executing the extrusion-lamination. The corona-treating conditions were as follows:

Corona-treating apparatus: Manufactured by Kasuga
Denki Co.

Treating condition: 40 W/m²/min.

Surface wet tension after the treatment: 58 dyns/cm

The results were as shown in Table 3.

(Example 9)

The laminated film was prepared according to the same method and under the same conditions as those of Example 1

but so preparing the resin composition of Example 1 that 18% of the carboxylic acid group of the unsaturated carboxylic acid was ionized with zinc and changing the temperature of the extrusion-laminated resin to 315 °C, and its physical properties were evaluated. The results were as shown in Table 4.

(Comparative Example 6)

The extrusion-lamination processing was executed according to the same method and under the same conditions as those of Example 1 but using a resin composition of (A-2) ethylene/methacrylic acid/isobutyl acrylate copolymer ② and (C-1) ethylene/ α -olefin copolymer ① blended at a ratio shown in Table 3 as a resin to be extrusion-laminated and changing the resin temperature to 315 °C. In this case, however, both ends of the molten resin film swung to a large extent during the extrusion-lamination processing and did not remain stable. Therefore, the laminated layer could not be sampled. The results were as shown in Table 3.

(Comparative Example 7)

The laminated film was prepared according to the same method and under the same conditions as those of Example 1 but using (B-2) ethylene/acrylic acid copolymer ② alone as a resin to be extrusion-laminated and changing the resin temperature to 310 °C. The obtained laminated film exhibited an adhesive force to the polyester film of as very low as 0.6 N/15 mm. The heat-sealing strength did not reach the practicable level, and the laminated film was judged to be not suited as a packaging material. The results were as shown in Table 3.

(Comparative Example 8)

Properties were evaluated according to the same method and under the same conditions as those of Example 1 but using a resin composition of (B-2) ethylene/acrylic acid copolymer ② and (C-2) ethylene/ α -olefin copolymer

② blended at a ratio shown in Table 3 as a resin to be extrusion-laminated and changing the resin temperature to 310 °C. The obtained laminated film exhibited an adhesive force to the polyester film of as low as 1.7 N/15 mm. The heat-sealing strength did not reach the practicable level, and the laminated film was judged to be not suited as a packaging material. The results were as shown in Table 3. (Comparative Example 9)

Properties were evaluated according to the same method and under the same conditions as those of Example 1 but using (D-1) polyethylene alone as a resin to be extrusion-laminated. The obtained laminated film exhibited an adhesive force to the polyester film of 2.6 N/15 mm which was slightly smaller than the target strength. The adhesive strength to the aluminum foil did not reach the practicable level, and the laminated film was judged to be not suited as a packaging material. (Comparative Example 10)

Properties were evaluated according to the same method and under the same conditions as those of Example 1 but using (E-1) ethylene/ethyl acrylate copolymer ① alone as a resin to be extrusion-laminated. The obtained laminated film exhibited an adhesive force to the polyester film of as very small as 0.9 N/15 mm. Besides, the adhesive strength to the aluminum foil was very small, and the laminated film was judged to be not suited as a packaging material. (Comparative Example 11)

The laminated film was prepared according to the same method and under the same conditions as those of Example 1 but so preparing the resin composition of Example 1 that 35% of the carboxylic acid based on the unsaturated carboxylic acid was ionized with zinc and changing the temperature of the extrusion-laminated resin to 315 °C, and its physical properties were evaluated. The results

were as shown in Table 4.

As will be obvious from Tables 1 to 3, it was confirmed that a laminate having a practically sufficient adhesive force to the polyester and heat-sealing property was obtained by extrusion-laminating the resin composition of the present invention onto a polyester film according to a predetermined extrusion-lamination processing method and under predetermined conditions. It was further confirmed that the resin composition of the invention exhibited excellent extrusion-lamination adhesiveness to the polar base member as represented by an aluminum foil, and was suited for laminating the polyester film with various polar base members.

15

20

25

30

35

T06220" 50E90250

Table 1

Laminated-resin composition (pts by wt.)		Example 1	Example 2	Comparative Example 1	Comparative Example 2	Comparative Example 3	Example 3
(A-1) Copolymer ①		50	50	50	50	50	
(A-2) Copolymer ②							100
(B-1) Ethylene/methacrylic acid copolymer ①		50	50	50	50	50	
(B-2) Ethylene/acrylic acid copolymer ②							
(C-1) Ethylene/ α -olefin copolymer ①							
(C-2) Ethylene/ α -olefin copolymer ②							
(C-3) Ethylene/ α -olefin copolymer ③							
(D-1) Polyethylene							
(E-1) Ethylene/ethyl acrylate copolymer							
Content of copolymer component in the laminated resin (% by wt.)		4	4	4	4	4	11
Unsaturated carboxylic acid (meth)acrylic acid ester		7.5	7.5	7.5	7.5	7.5	8

Table 1 (continued)

	Example 1	Example 2	Comparative Example 1	Comparative Example 2	Comparative Example 3	Example 3
<u>Extrusion-laminated resin temp. (°C)</u>	300	320	270	350	320	300
<u>Surface wet tension of laminated surface of polyester film (dyns/cm)</u>	56	56	56	56	44	56
<u>Extrusion-lamination processability</u>						
Max. Processing rate (m/min)	400<	400<	300		400<	400<
Condition of laminated film	○	○	○	×	○	○
<u>Adhesive force to polyester film (N/15 mm)</u>	6.9	10.6	0.1<	could not be evaluated	0.8	4.6
<u>Adhesive force to aluminum foil (N/15 mm)</u>	2.7	2.8	2.8	could not be evaluated	2.8	2.8
<u>Heat-sealing strength (N/15 mm)</u>						
120 °C	35	37	12	could not be evaluated	14	39
140 °C	36	38	14	could not be evaluated	16	40
160 °C	36	38	13	could not be evaluated	16	41

Table 2

Laminated-resin composition (pts by wt.)	Table 2				
	Example 4	Example 5	Comparative Example 4	Example 6	Example 7
(A-1) Copolymer ①					45
(A-2) Copolymer ②					45
(B-1) Ethylene/methacrylic acid copolymer ①	90	90	90	90	
(B-2) Ethylene/acrylic acid copolymer ②					45
(C-1) Ethylene/ α -olefin copolymer ①	10				10
(C-2) Ethylene/ α -olefin copolymer ②		10	10		
(C-3) Ethylene/ α -olefin copolymer ③				10	
(D-1) Polyethylene					
(E-1) Ethylene/ethyl acrylate copolymer					
Content of copolymer component in the laminated resin (% by wt.)					
Unsaturated carboxylic acid	9.9	9.9	9.9	9.9	3.6
(meth)acrylic acid ester	7.2	7.2	7.2	7.2	6.8

Table 2 (continued)

Example 4	Example 5	Comparative Example 4	Example 6	Example 7	Comparative Example 5
<u>Extrusion-laminated resin temp. (°C)</u>					
285	300	300	300	315	315
<u>Surface wet tension of laminated surface of polyester film (dyns/cm)</u>					
56	56	44	56	56	44
<u>Extrusion-lamination processability</u>					
400<	400<	400<	370	400<	400<
○	○	○	○	○	○
<u>Condition of laminated film</u>					
3.5	7.1	0.7	5.4	9.4	0.6
<u>Adhesive force to polyester film (N/15 mm)</u>					
2.6	2.8	2.8	2.8	2.8	2.8
<u>Adhesive force to aluminum foil (N/15 mm)</u>					
37	not evaluated	15	not evaluated	not evaluated	not evaluated
39	not evaluated	17	not evaluated	not evaluated	not evaluated
39	not evaluated	16	not evaluated	not evaluated	not evaluated
<u>Heat-sealing strength (N/15 mm)</u>					
120 °C					
140 °C					
160 °C					

Table 3

Laminated-resin composition (pts by wt.)		Example 8	Comparative Example 6	Comparative Example 7	Comparative Example 8	Comparative Example 9	Comparative Example 10
(A-1) Copolymer ①							
(A-2) Copolymer ②		90	65				
(B-1) Ethylene/methacrylic acid copolymer ①							
(B-2) Ethylene/acrylic acid copolymer ②			100		90		
(C-1) Ethylene/ α -olefin copolymer ①			35				
(C-2) Ethylene/ α -olefin copolymer ②		10			10		
(C-3) Ethylene/ α -olefin copolymer ③						100	
(D-1) Polyethylene							
(E-1) Ethylene/ethyl acrylate copolymer						100	
Content of copolymer component in the laminated resin (% by wt.)							
Unsaturated carboxylic acid		9.9	7.2	5	4.5	0	0
(meth)acrylic acid ester		7.2	5.2	0	0	0	9

Table 3 (continued)

	Example 8	Comparative Example 6	Comparative Example 7	Comparative Example 8	Comparative Example 9	Comparative Example 10
<u>Extrusion-laminated resin temp. (°C)</u>	300	315	310	310	300	300
<u>Surface wet tension of laminated surface of polyester film (dyns/cm)</u>	58	56	56	56	56	56
<u>Extrusion-lamination processability</u>						
Max. Processing rate (m/min)	370		400<	350	200	220
Condition of laminated film	○	×	○	○	○	○
<u>Adhesive force to polyester film (N/15 mm)</u>	7.9	could not evaluated	0.6	1.7	2.6	0.9
<u>Adhesive force to aluminum foil (N/15 mm)</u>	not evaluated	could not evaluated	3.7	3.5	1.6	0.3
<u>Heat-sealing strength (N/15 mm)</u>						
120 °C	not evaluated	could not be evaluated	18	18	not evaluated	not evaluated
140 °C	not evaluated	could not be evaluated	20	19	not evaluated	not evaluated
160 °C	not evaluated	could not be evaluated	19	17	not evaluated	not evaluated

Table 4

<u>Laminated-resin composition (pts by wt.)</u>	<u>Example</u> <u>9</u>	<u>Comparative</u> <u>Example 11</u>
(A-1) Copolymer ①	50	50
(B-1) Ethylene/methacrylic acid copolymer ①	50	50
<u>Content of copolymer component in the laminated resin</u> <u>(% by wt.)</u>		
Unsaturated carboxylic acid	4	4
(meth)acrylic acid ester	7.5	7.5
<u>Ratio of ionized unsaturated carboxylic acid (%)</u>	18	35
<u>Extrusion-laminated resin temp. (°C)</u>	315	315
<u>Surface wet tension of laminated surface of polyester film</u> <u>(dyns/cm)</u>	56	56
<u>Extrusion-lamination processability</u>		
Max. Processing rate (m/min)	400<	400<
Condition of laminated film	○	○
<u>Adhesive force to polyester film (N/15 mm)</u>	6.8	2.8
<u>Adhesive force to aluminum foil (N/15 mm)</u>	2.6	2.4
<u>Heat-sealing strength (N/15 mm)</u>		
120 °C	33	20
140 °C	34	22
160 °C	35	23

According to the present invention, it is allowed to obtain a laminated film having excellent extrusion processability and exhibiting a large interlayer adhesive force upon being extrusion-laminated directly on the polyester film. Even when sandwich-laminated together with a polar base member, the laminated film exhibits good interlayer adhesive force to the polar base member. Besides, there is no need of executing the anchor-coat processing for the polyester film offering advantage from the standpoint of maintaining sanitary environment. Moreover, since the molten resin film of the extrusion-laminated resin needs not be treated with ozone, the packaging material can be produced at a low cost, offering a great merit for the manufacturers of the packaging materials. The thus obtained laminated film can be extensively used as a variety of packaging members for packaging foods such as cakes, daily dishes, beverages, processed meat, etc. which are packaged by using polyester films.

20

25

30

35

T062E0" 50E90860

CLAIMS

1. A laminated film obtained by extrusion-laminating, onto at least one surface of a polyester film,
5 an ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer or a mixture resin composition thereof with an ethylene/unsaturated carboxylic acid copolymer and/or an ethylene/(meth)acrylic acid ester copolymer, the amount of the unsaturated carboxylic acid component being
10 from 1 to 12% by weight and the amount of the (meth)acrylic acid ester component being from 2 to 25% by weight with respect to the total amount of said extrusion-laminated resin components.

2. A laminated film obtained by extrusion-laminating, onto at least one surface of a polyester film,
15 a mixture resin comprising:

(a) 100 parts by weight of the mixture resin component of an ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer or the mixture
20 resin composition of said ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer with an ethylene/unsaturated carboxylic acid copolymer and/or an ethylene/(meth)acrylic acid ester copolymer, the amount of the unsaturated carboxylic acid component being from 1 to
25 12% by weight and the amount of the (meth)acrylic acid ester component being from 2 to 25% by weight with respect to the total amount of the extrusion-laminated resin components, and

(b) not more than 30 parts by weight of an ethylene/
30 α -olefin copolymer resin having a density of 840 to 900 kg/m³.

3. A laminated film according to claim 1, wherein the carboxylic acid group of the unsaturated carboxylic acid in said mixture resin composition is partly ionized
35 with an alkali metal or an alkaline earth metal within a

range in which the ionization degree is not larger than 20%.

4. A laminated film according to claim 2, wherein the carboxylic acid group of the unsaturated carboxylic acid in said mixture resin is partly ionized with an alkali metal or an alkaline earth metal within a range in which the ionization degree is not larger than 20%.

5. A laminated film according to claim 1 or 2, wherein another polar base member is laminated on the polyester film via the extrusion-laminated resin.

6. A method of producing a laminated film by extrusion-laminating, on the surface of a polyester film of which the surface is oxidized and has a surface wet tension of not smaller than 45 dyns/cm, (1) an extrusion-lamination resin: an ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer or a mixture resin composition thereof with an ethylene/unsaturated carboxylic acid copolymer and/or an ethylene/(meth)acrylic acid ester copolymer, the amount of the unsaturated carboxylic acid component being from 1 to 12% by weight and the amount of the (meth)acrylic acid ester component being from 2 to 25% by weight with respect to the total amount of said extrusion-laminated resin components, or an ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer, or (2) an extrusion-lamination resin: a mixture resin comprising:

(a) 100 parts by weight of the mixture resin component of an ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer or the mixture resin composition of said ethylene/unsaturated carboxylic acid/(meth)acrylic acid ester copolymer with an ethylene/unsaturated carboxylic acid copolymer and/or an ethylene/(meth)acrylic acid ester copolymer, the amount of the unsaturated carboxylic acid component being from 1 to 12% by weight and the amount of the (meth)acrylic acid

ester component being from 2 to 25% by weight with respect to the total amount of the extrusion-laminated resin components, and

(b) not more than 30 parts by weight of an ethylene/
5 α -olefin copolymer resin having a density of 840 to 900 kg/m³, at a resin temperature of from 280 to 340 °C.

7. A method of producing a laminated film according to claim 6, wherein said extrusion-lamination resin is extrusion sandwich-laminated between said polyester film
10 and another polar base member.

15

20

25

30

35

T06220" 50E90860

ABSTRACT

A laminated film exhibiting excellent interlayer adhesiveness obtained by extrusion-laminating a resin
5 directly on a polyester film without effecting anchor-coating or without treating the molten film of the extrusion-laminated resin with ozone. The laminated film is obtained by extrusion-laminating, onto the polyester film, an ethylene copolymer containing an unsaturated
10 carboxylic acid and a (meth)acrylic acid ester, which may be further blended with a small amount of an ethylene/ α -olefin copolymer having a density of 840 to 900 kg/m³, the amount of the unsaturated carboxylic acid component being from 1 to 12% by weight and the amount of the
15 (meth)acrylic acid ester component being from 2 to 25% by weight.

20

25

30

35

T.05220" 50E90350

Dkt. No. SPO-593**DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (*if only one name is listed below*) or an original, first and joint inventor (*if plural names are listed below*) of the subject matter which is claimed and for which a patent is sought on the invention entitled
Laminated film and method of producing the same_____, the specification of which:
(check one) ☒ is attached hereto ☐ was filed on _____ as
Application Serial No. _____ and
was amended on _____
(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT international application which designated at least one country other than the United States, listed below and have also identified below any foreign application for patent or inventor's certificate, or PCT international application having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)			Priority Claimed
<u>231527/99</u>	<u>Japan</u>	<u>18/8/1999</u>	<input checked="" type="checkbox"/> <input type="checkbox"/>
(NUMBER)	(COUNTRY)	(FILED D/M/Y)	YES NO
_____	_____	_____	<input type="checkbox"/> <input type="checkbox"/>
(NUMBER)	(COUNTRY)	(FILED D/M/Y)	YES NO

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

<u> </u>	<u> </u>
(APPLICATION NUMBER)	(FILING DATE)
<u> </u>	<u> </u>
(APPLICATION NUMBER)	(FILING DATE)

I hereby claim the benefit under 35 U.S.C. §120 of any United States application(s), or §365(c) of any PCT international application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of 35 U.S.C. §112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

<u>PCT/JP00/03904</u>	<u>June 15, 2000</u>	<u>pending</u>
(APPLICATION SERIAL NO.)	(FILING DATE)	(STATUS)
<u> </u>	<u> </u>	<u> </u>
(APPLICATION SERIAL NO.)	(FILING DATE)	(STATUS)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorneys and/or agents to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

Leonard W. Sherman	Reg. No. <u>19,636</u>	Alan Holler	Reg. No. <u>29,266</u>
Edwin A. Shalloway	Reg. No. <u>19,967</u>	Karl Hoback	Reg. No. <u>23,026</u>
Richard A. Steinberg	Reg. No. <u>26,588</u>	Robert L. Haines	Reg. No. <u>35,533</u>
Perry Carvellas	Reg. No. <u>19,637</u>		

SEND CORRESPONDENCE TO:

DIRECT TELEPHONE CALLS TO:

SHERMAN & SHALLOWAY
413 North Washington Street
Alexandria, Virginia 22314

(703) 549-2282

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine and imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or document or any patent issuing thereon.

Full name of sole or first inventor Kenji Miharu
GIVEN NAME MIDDLE NAME FAMILY NAME
Inventor's signature Kenji Miharu
Date of signature March 20, 2001
Residence Chiba JPX Chiba-ken Japan
CITY STATE OR PROVINCE COUNTRY
Citizenship Japan
Post Office Address 1-17-24, Miyazaki, Chuou-ku, Chiba-shi,
(insert complete mailing address, including country) Chiba-ken, Japan

Full name of second inventor Naozumi Suzuki
GIVEN NAME MIDDLE NAME FAMILY NAME
Inventor's signature Naozumi Suzuki
Date of signature March 20, 2001
Residence Ichihara JPX Chiba-ken Japan
CITY STATE OR PROVINCE COUNTRY
Citizenship Japan
Post Office Address 4-8-6, Minamikokubunjidai, Ichihara-shi,
(insert complete mailing address, including country) Chiba-ken, Japan

Full name of third inventor _____
GIVEN NAME MIDDLE NAME FAMILY NAME
Inventor's signature _____
Date of signature _____
Residence _____
CITY STATE OR PROVINCE COUNTRY
Citizenship _____
Post Office Address _____
(insert complete mailing address, including country) _____

☐ ADDITIONAL INVENTORS ARE BEING NAMED ON SEPARATELY NUMBERED SHEETS ATTACHED HERETO